Size and mass of Cooper pairs determined by low-energy μSR and PNR

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The Pippard coherence length ξ_0 (the size of a Cooper pair) in an extreme type-I superconductor was determined *directly* through high-resolution measurement of the nonlocal electrodynamic effect combining low-energy muon spin rotation spectroscopy and polarized neutron reflectometry. The renormalization factor $Z \equiv m^*_{cp}/2m$ (m^*_{cp} and m are the mass of the Cooper pair and the electron, respectively) resulting from the electron-phonon interaction, and the temperature dependent London penetration depth $\lambda_{\rm L}(T)$ were determined as well. A general expression linking ξ_0, Z and $\lambda_{\rm L}(0)$ is introduced and experimentally verified. This expression allows one to determine experimentally the Pippard coherence length in *any* superconductor, independent of whether the electrodynamics is local or nonlocal, conventional or unconventional.

PACS numbers:

A core concept of superconductivity is Cooper pairing of electrons. Cooper pairs have a characteristic size ξ_0, the Pippard coherence length, and an effective mass $m^*_{cp} \equiv 2Zm$, m being the Coulomb and band-structure effective mass of electron and Z the renormalization factor, a measure of the electron-boson coupling strength [1]. Only weakly dependent on temperature T, ξ_0 provides a reference length for the fundamental length scales, including in particular the London penetration depth $\lambda_{\rm L}(T)$, characterizing the decay of a penetrating magnetic field. In units of ξ_0, $\lambda_{\rm L}(T)$ determines whether the superconductor is described by *local* or *nonlocal* electrodynamics, and $\lambda_{\rm L}(0)$ determines whether it is type-I or type-II [2]. To ensure consistency ξ_0 and Z should be measured simultaneously. However, in spite of their importance, ξ_0 and Z have not yet been directly and simultaneously measured in any superconductor.

As shown by Pippard [3], ξ_0 can be determined in *nonlocal* superconductors through measurement of the nonlocal electrodynamic effect [4]. Most superconductors are local and for those it was stated that ξ_0 cannot be measured directly [5]. Here we report on a direct and simultaneous determination of ξ_0, Z and $\lambda_{\rm L}$ in a nonlocal superconductor (In) and answer the question how ξ_0 can be determined experimentally in any superconductor.

When a superconductor is in the Meissner state, an external magnetic field B_0 is completely screened due to a persistent current running in a thin surface layer over which the field decays to zero. The layer thickness is of the order of the "magnetic penetration depth" $\lambda \equiv B_0^{-1}\int_0^\infty B(z)dz$, z being the distance from the surface. If the size of the Cooper pairs is small ($\xi_0 \ll \lambda$), the relationship between the current density and the vector potential can be treated as local. Then the field decays as $\exp(-z/\lambda_{\rm L})$, where $\lambda_{\rm L} = \sqrt{\Lambda c^2/4\pi}$ is the London penetration depth (c is the speed of light). In the London theory the phenomenological parameter Λ is a function of the mass and the density of superconducting electrons, none of the two being well defined [2].

The local approximation is applicable to superconductors with Ginzburg-Landau parameter $\kappa \gtrsim 1.6$ [6]. For superconductors with smaller κ the Cooper pair size is no longer negligible and the current density is determined by the vector potential averaged over a region of dimension ξ_0. The nonlocality leads to deeper field penetration and to distortion of the $B(z)$ shape. In the pure limit (elastic mean free path $\ell \gg \xi_0$) $B(z)$ is a function of both $\lambda_{\rm L}(T)$ and ξ_0. Therefore, knowledge of $B(z)$ in low-κ materials allows one to determine these two parameters.

As predicted by Pippard and supported by the Bardeen-Cooper-Schrieffer (BCS) theory, in nonlocal superconductors $B(z)$ is nonmonotonic with a sign reversal at a certain depth. This *nonlocal electrodynamic effect* is most pronounced in extreme type-I superconductors, such as In ($\kappa \approx 0.07$); it can also occur in unconventional superconductors with nodes in the energy gap [5].

The nonlocal effect has been directly confirmed by low-energy muon spin rotation (LE-μSR) measurements on Pb, Ta and Nb [7, 8], and soon after by measurements of polarized neutron reflectivity (PNR) in In [9]. In Refs. [7, 8] a first attempt was made to infer ξ_0 from $B(z)$ in Pb. However, due to an unresolved issue of systematic errors only statistical errors were estimated. One can resolved this issue combining LE-μSR and PNR measurements since in the latter the systematic errors can be excluded.

The knowledge of $B(z)$ in nonlocal superconductors

also allows one to determine Z. Electrons near the Fermi surface are dressed by a cloud of virtual phonons, leading to an enhancement of their effective mass and consequently to a reduction of the Fermi velocity $v_{\rm F}$. Below the critical temperature T_c the phonon mediated attraction of elections exceeding their screened Coulomb repulsion results in formation of the Cooper pairs. In BCS theory electron-electron coupling is weak: $N(0)V \ll 1$, where V is the pairing potential and $N(0)$ is the single-spin electron density of states at the Fermi surface obtained from specific-heat measurements. Eliashberg's strong-coupling theory (SCT) is free from this limitation and agrees better with experiments [1].

In SCT the effective mass of electrons near the Fermi surface is $m^* = Zm$ and consequently the effective mass of the Cooper pairs is $m^*_{cp} = 2Zm$. Correspondingly, $\lambda_{\rm L}$ and ξ_0 are renormalized with respect to their values in the weak-coupling (wc) limit as

$$\lambda_{\rm L} = \sqrt{Z}\ \lambda_{\rm L}^{wc}, \qquad (1)$$

$$\xi_0 = \xi_0^{wc}/Z. \qquad (2)$$

If $\lambda_{\rm L}(T \to 0)$ is measured, the factor Z can be inferred from Eq. (1). $\lambda_{\rm L}^{wc}(0)$ can be obtained from the Faber-Pippard formula for Λ derived for the electrons not interacting with the lattice [10]. This leads to

$$\lambda_{\rm L}^{wc}(0) = \sqrt{\frac{3c^2}{8\pi e^2 N(0) v_F^2}}\ , \qquad (3)$$

where e is the electron charge. On the other hand, if ξ_0 is measured, Z can be calculated from Eq. (2) using the BCS definition $\xi_0^{wc} \equiv \hbar v_{\rm F}/\pi\Delta(0) = 0.18\hbar v_{\rm F}/k_B T_c$, where $\Delta(0) \equiv \Delta(T = 0)$ is the energy gap and $h = \hbar \times (2\pi)$ and k_B are the Planck and Boltzmann constant, respectively.

Unfortunately neither of these approaches is applicable for quantitative analysis since reliable calculation of $v_{\rm F}$ is not possible due to the complex topology of the Fermi surface in polyvalent metals [11]. However, if both $\lambda_{\rm L}(0)$ and ξ_0 are known, one can eliminate $v_{\rm F}$, thus obtaining

$$Z = \frac{c^2\hbar^2}{12.5\pi T_c^2 e^2 \gamma} \cdot \frac{1}{\lambda_{\rm L}(0)^2 \xi_0^2}\ , \qquad (4)$$

where it is taken into account that $N(0) = 3\gamma/2\pi^2 k_B^2$ (γ is the electron heat capacity coefficient [11]). Eq. (4) is a *general expression* independent of the relationship between the current density and the vector potential and of the specific nature of the electron-electron pairing.

In SCT $Z = 1 + \lambda_m$, where λ_m is a mass-enhancement parameter calculated in Ref. [1] using tunneling spectroscopy experimental data. λ_m can also be obtained from McMillan's equation for T_c [12], but this approach is less reliable [1]. Therefore, for nonlocal superconductors Eq. (4) furnishes a bridge between the $B(z)$ and the tunneling data, hence providing an independent test for the validity of $\lambda_{\rm L}$ and ξ_0 inferred from the $B(z)$ data. On the other hand, the vast majority of superconductors are local. For those Eq. (4) allows one to determine ξ_0 from the measured values of $\lambda_{\rm L}(0)$ and Z using, e.g., LE-μSR and tunneling experiments, respectively.

TABLE I: Parameters of the samples. d is the thickness, o is the oxide layer thickness (see Ref. [9]), RRR is the residual resistivity ratio; ℓ is the elastic mean free path.

sample	T_c(K)	size (cm)	d (μm)	o (nm)	RRR	ℓ (μm)
IN-1	3.415	2×3	2.5	≤1	560	11
IN-2	3.415	Ø 6	3.3	≤1	730	14

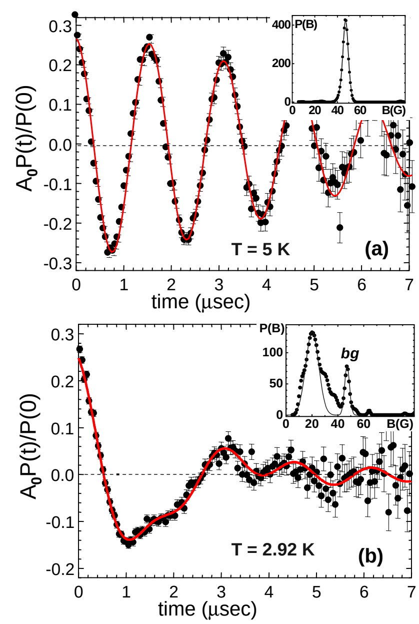

FIG. 1: Time spectra of muons with energy of 22 keV at an applied field of 47 Oe in the normal state (a) and in the Meissner state (b). The curves are fits to the Gaussian (a) and the time-domain (b) models. The inserts present the field spectra $P(B)$. In the insert in (b) bg is the background contribution, while the curve represents two Gaussian peaks to approximate the experimental spectrum. A_0 is the maximum observable asymmetry of the muon decay.

In this work $B(z)$ is measured using the LE-μSR [13, 14] and PNR [15] techniques. Combining these complementary techniques enables an independent verification of the inferred values of ξ_0 and $\lambda_{\rm L}(T)$ and an estimate of there total uncertainties. The samples were two indium films whose parameters are listed in table 1. The films were deposited on an oxidized silicon wafer (IN-1) and on a sapphire crystal (IN-2) held at room temperature via thermal evaporation of indium shots (99.9999% purity) at a base pressure $\lesssim 5 \cdot 10^{-9}$ mbar. The IN-1 sample was used for PNR measurements and for LE-μSR measurements down to 2.9 K. The IN-2 sample was used for LE-μSR measurements at lower temperatures.

The samples' DC magnetization exhibits a clear-cut first-order phase transition with deep supercooling. The

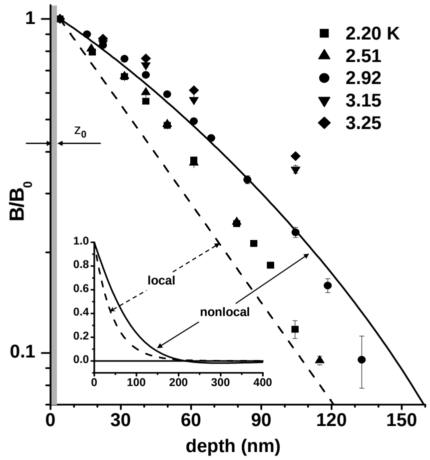

FIG. 2: Reduced field B/B_0 obtained from the LE-μSR measurements using the Gaussian model. The depth is the average stopping distance of the implanted muons calculated from the Monte Carlo TRIM.SP code. The solid (dashed) line is the field profile calculated from the nonlocal (local) theory at T = 2.92 K with ξ_0=380 nm, $\lambda_{\rm L}(0)$=30 nm and the dead layer z_0= 4 nm, inferred from the χ^2 and the time-domain analysis. The insert presents the same field profiles on a linear scale.

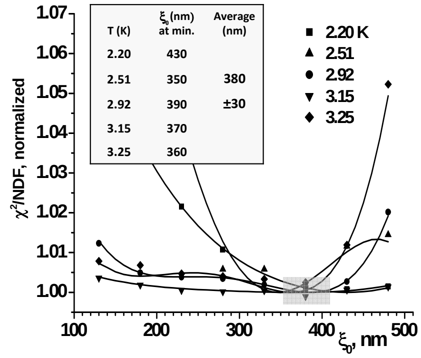

FIG. 3: χ^2/NDF versus ξ_0 and the corresponding polynomial fittings normalized to the minimum value for each temperature. The shaded area indicates the range of the best estimates for ξ_0. The inserted table gives positions of the minima and their average with the standard deviation.

measured critical field $H_c(T)$ and T_c perfectly agree with values reported in literature [16, 17]. The surface of the films consists of nearly atomically flat terraces (root-mean-square roughness of the terrace areas is $\lesssim 2$ Å) with a typical size around 5 μm with voids in between. This size is much larger than ξ_0 and the total area of the voids does not exceed 3% of the sample surface. Therefore the terrace surface structure should not affect the electrodynamic properties of the films. For both samples the elastic mean free path $\ell >> \xi_0$, therefore the samples are type-I superconductors in the pure limit [2].

The **LE-μSR experiments** were performed at the μE4 beamline of the Swiss Muon Source at the Paul Scherrer Institute [18]. Typical spectra of muon polarization P are presented in Fig. 1. In the normal state B is uniform and the $P(B)$ spectrum (insert in Fig. 1a) has Gaussian shape due to nuclear dipole broadening. In the Meissner state B is not uniform and $P(B)$ is not Gaussian. However, Gaussian spectra (curve in the insert of Fig. 1b) provide a good first approximation.

$B(z)$ points obtained using a Gaussian model [8] for the depolarization of the precessing muons are presented in Fig. 2 along with the $B(z)$ curve calculated from the local and nonlocal theories . The sample temperature was determined *in situ* based on the $H_c(T)$ phase diagram obtained from magnetization measurements. As can be seen in Fig. 2, the "Gaussian" B-points exhibit a pronounced non-exponential depth dependence, consistent with the nonlocal effect. Discrepancies between the points and the "nonlocal" theoretical curve are mainly caused by incomplete adequacy of the Gaussian model. On the other hand, the qualitative consistency of the "Gaussian" points with the nonlocal theory justifies the application of a time-domain model [19], directly assuming the "nonlocal" shape of $B(z)$ with $\lambda_{\rm L}$ as an adjustable parameter.

In nonlocal superconductors the effective penetration depth $\lambda_{\rm eff} \approx (\lambda_{\rm L}^2\xi_0)^{\frac{1}{3}}$ [2]. Therefore either $\lambda_{\rm L}$ or ξ_0 has to be determined using an additional criterium. Since $\lambda_{\rm eff}$ is mainly sensitive to $\lambda_{\rm L}$ and ξ_0 weakly depends on temperature, this parameter is ξ_0. Its optimal value was found from χ^2 analysis of the global fits (simultaneous fitting of all implantation energy data sets at each temperature). The fits were performed with different ξ_0 chosen around a theoretical value of 377 nm [12]. The graphs for χ^2/NDF (NDF stands for number of degrees of freedom) versus ξ_0 are presented in Fig. 3. Values of ξ_0 at the minima and the best estimate for ξ_0 are given in the insert. The values of $\lambda_{\rm L}$ obtained from the global fits with $\xi_0 = 380$ nm are presented in a summary graph in Fig. 5.

The **PNR measurements** were performed on the D3 reflectometer at the NRU reactor in Chalk River. The experimental data and simulations for the reflectivity of neutrons polarized parallel (R^+) and antiparallel (R^-) to the magnetic field and for the spin asymmetry $(R^+ - R^-)/(R^+ + R^-)$ are presented in Fig. 4. In agreement with our LE-μSR results and previous PNR results [9], the neutron spin asymmetry (Fig. 4, insert (a)) simulated with $B(z)$ calculated from the nonlocal theory matches the experimental data significantly better than the simulation based on the London field profile. The "nonlocal" $B(z)$ was calculated with $\xi_0 = 380$ nm obtained from the LE-μSR data. The best match of the simulated spin asymmetry with the experimental data was achieved for $\lambda_{\rm L}(T = 0.3\,{\rm K}) = 28.0 \pm 2.5$ nm.

Results for $\lambda_{\rm L}$ inferred from the LE-μSR and PNR measurements are shown in Fig. 5. The values of $\lambda_{\rm L}(T)$ are consistent with each other and agree with the two-fluid formula [2]. The best estimate of $\lambda_{\rm L}(0)$ is 30 ± 2 nm.

Having determined $\lambda_{\rm L}(0)$ and ξ_0 we calculated Z from Eq. (4). The results are summarized in the table inserted in Fig. 5, from which it can be concluded that the value of

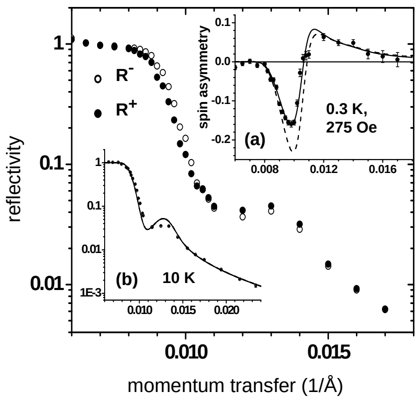

FIG. 4: Neutron reflectivity in the Meissner state at $T = 0.3\,\mathrm{K}$ and $B_0 = 275\,\mathrm{Oe}$. Insert (a): Measured spin asymmetry (points) and its simulation with $B(z)$ obtained from the local (dashed curve) and nonlocal (solid curve) theories with $\lambda_{\mathrm{L}} = 28\,\mathrm{nm}$ and $\xi_0 = 380\,\mathrm{nm}$. Insert (b): Measured (points) and simulated (curve) reflectivity in the normal state.

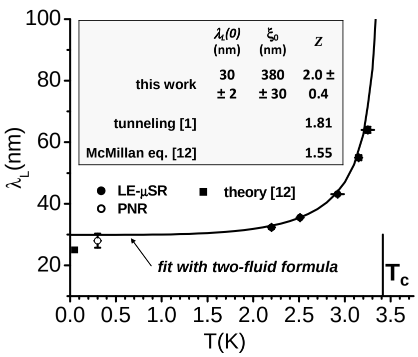

FIG. 5: Temperature dependence of the London penetration depth in indium. The inserted table gives the values of $\lambda_{\mathrm{L}}(T \to 0)$, ξ_0 and Z obtained in this work, and the values of Z calculated from the tunneling spectroscopy data [1] and from McMillan's equation for T_c [12].

Z obtained in this work agrees with the value obtained from electron tunneling data. The latter implies that our results *quantitatively* confirm the nonlocal electrodynamic effect and *confirm* the validity of Eq. (4).

In conclusion, *high resolution* measurements of the magnetic field profiles $B(z,T)$ were performed, for the first time, in an extreme type-I superconductor (indium) combining LE-μSR and PNR measurements. The $B(z)$ profiles *quantitatively* confirm the Pippard/BCS nonlocal electrodynamic effect. The $B(z)$ data were used to determine the *Pippard coherence length* ξ_0, the *renormalization factor* Z for the electron-phonon mass enhancement of the Cooper pairs, and the *London penetration depth* $\lambda_{\mathrm{L}}(T)$. The experimentally verified equation (4) allows one to infer ξ_0 in *any* superconductor from the results for $\lambda_{\mathrm{L}}(0)$ and Z.

Acknowledgments. We thank Tom Moorkens, Maarten Trekels and Bastian Wojek for help with the measurements. This work was supported by the National Science Foundation (DMR 0904157) and by the Science Foundation - Flanders (FWO); J.O.I. acknowledges the support by KU Leuven grant OT/11/063.

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